

We would like to thank reviewer 2 for the effort, interesting questions, and constructive comments. Below we address the points one by one. The reviewer’s comments are listed in cursive, with our answers in blue and excerpts from the revised text in red.

*Summary: This study aims to provide global CO emission estimates for late 2018, particularly in the northern hemisphere, using a top-down inverse modeling approach. By incorporating TROPOMI satellite observations into the TM5-4DVAR model and further constraining emissions with NOAA surface flask measurements, six experiments were conducted to assess the impact of different emissions and observational datasets on inversion outcomes. The main findings of this paper are: 1) The satellite-only inversion closely matches flask measurements south of 55°N, suggesting suitability for real-time applications; 2) Up to 75% emission reductions in China and India, with reductions in China attributed to policy changes; 3) Outstanding issues were identified to include underestimation of OH causing lower emissions, and localized emission increments over Europe and the Sahara.*

*Main Comments: While CO inversions have been conducted for over 20 years, there are still gaps in our understanding of CO sources and sinks despite availability of satellite data. This makes this paper potentially a relevant contribution to the scientific community and this journal given the importance of CO in understanding atmospheric composition. The authors have also described their inverse methodology in quite (and understandably) in detail which potentially can enable easier connection to relevant findings and issues identified in making the inverse framework suitable for near-real time applications. However, several concerns with this manuscript require revision:*

*1. The title “Efficacy of High-Resolution Satellite Observations” may be misleading, as approximations (e.g., model resolution and spatiotemporal scales of inversion) challenge this claim, and the findings don’t fully support it.*

We agree with this assessment. The title has been revised to:

“Top-down CO emission estimates using TROPOMI CO data in the TM5-4DVAR (r1258) inverse modeling suit”

*2. While the approach shows promise in reducing uncertainties compared to flask measurements, its suitability for near real-time inversions is not adequately demonstrated in this manuscript.*

Discussions regarding a near-real-time setup as a longer-term goal had been removed in a previous iteration of the manuscript. We apologize for any confusion caused by the remaining references in the manuscript that were now removed.

*3. Verification is limited, as there’s no comparison with other satellite observations or top-down emission estimates for this period. Although TROPOMI offers extensive coverage, it has limitations due to its SWIR-only focus; integrating IASI, MOPITT, and CrIS would better constrain synoptic CO patterns and widespread anthropogenic CO, which flask data alone cannot capture. One way to alleviate this are: a) comparison with other top-down estimates, b) to make objectives to be more focused.*

We thank the reviewer for this useful suggestion. As mentioned in the response to the third comment of the first reviewer (RC1), this manuscript focuses primarily on TROPOMI data and flask observations. For additional details, please see our response there (AC1). However, following the reviewer’s suggestion, we have rewritten the objective to more clearly set our study apart from previous studies found in the literature and added a more in-depth comparison of the budget terms found by our inversion to the ones from other inversion studies. The objective at the end of the Introduction (L105ff) now reads:

“In this study, we investigate the added value of the new TROPOMI data for constraining global CO emissions in the TM5-4DVAR inverse modeling suit. Previous studies have already investigated the efficacy of TROPOMI observations for constraining the global atmospheric CO abundance (Inness et al., 2022) or CO emissions at regional to sub-city scales (Borsdorff et al., 2019, 2020; Sun, 2022; Tian et

al., 2022; Shahrokhi et al., 2023). Our study provides global CO emission estimates with a focus on the northern hemisphere in the second half of 2018. In addition to introducing TROPOMI observations into TM5-4DVAR, we have updated several input datasets, including the a priori emissions, and improved the methodology for handling satellite observations, most notably the weighting of multiple observational datasets in inversions, compared to previous studies using TM5-4DVAR (e.g. Krol et al., 2013; Nechita-Banda et al., 2018; Naus et al., 2022). We have divided the investigation of all of these changes into a series of experiments, in which we run the same inversion multiple times, each time with slightly different settings.”

The added comparison with other top-down estimates can be found in L743ff and reads:

“Overall, the a posteriori secondary CO source is lower than the a priori production flux in all experiments, as can be seen in the global budgets provided in Table 3, where the posterior masses at the end of the inversion period (final masses) are consistently lower than the prior final masses. Naus et al. (2022), who used a similar setup, also found too high secondary CO production. All fluxes were extrapolated to annual budget terms in  $\text{Tg CO yr}^{-1}$ . This extrapolation may lead to misleading results when compared to budget terms published elsewhere, because the inversion period of the main inversions includes the biomass burning season, but excludes the increased anthropogenic emissions due to heating during most of the northern hemispheric winter. With this caveat in mind, we compare our prior and posterior budget terms with values from other inversion studies with different setups, namely to Jiang et al. (2017), who assimilated MOPITT CO and methyl chloroform surface measurements in the GEOS-Chem model, Müller et al. (2018), who assimilated IASI CO in the IMAGES model, and Zheng et al. (2019), who assimilated MOPITT CO in the LMDz-SACS model. A detailed comparison of these three studies can be found in Elguindi et al, (2020). Compared to the results of either of those studies, our extrapolated annual a priori budget terms for secondary CO production and chemical loss of CO to OH are far too large. However, our posterior chemical loss falls between the values found in Müller et al. (2018) and Zheng et al. (2019) and our posterior secondary CO production, while still larger, is much closer to what those studies found than our prior. This improved agreement implies that our a posteriori terms are more realistic than the a priori ones. Note that our secondary production implicitly includes ocean and biogenic CO. While the total production and loss terms show reasonably good agreement with the aforementioned studies, the partitioning by source category of our emission terms differs slightly. Our anthropogenic/fossil fuel a posteriori CO is close to that found by Müller et al. (2018) and Jiang et al. (2017), but significantly lower than that reported by Zheng et al. (2019). In contrast, our biomass burning estimate is close to the multi-year mean of Zheng et al. (2019). However, due to the high year-to-year variability in biomass burning emissions, as shown by both Müller et al. (2018) and Zheng et al. (2019), this result is difficult to interpret, especially since neither study covers 2018.”

4. *Methodology presentation could be clarified for readers less familiar with inverse modeling.*

While we tried to implement further clarifications, additional comments as to which parts are still unclear are welcome.

5. *Enhance overall clarity, particularly by: a) clearly articulating key gaps in CO inversion methodology and current top-down estimates, and b) deepening the discussion of noteworthy findings that are better supported by experiments.*

The introduction has been extended to better contextualize our manuscript within the existing body of literature, as detailed in the response to comment 3. above.

*Specific Comments:*

1) *Recommend changing the title to reflect main findings supported by experiments.*

See response to comment 1. above.

2) *While there is a good description of the methods, it may be more clear and easy to follow with an addition of a table listing all key approximations (resolution of observations and models, inflation pa-*

rameters, super-observation parameters, inversion spatial and temporal windows, spin-up, specification of errors incl. length scales).

The requested information has been aggregated in a new supplementary Table S1, with the inflation factors added to Table 1.

3) *Abstract: While the length constraints are understood, several statements need clarification for clarity and impact:*

a) *What is the significance of 55°N, and why would capturing measurements below this latitude suggest suitability for near real-time inversions?*

Reference to near real-time inversions has been removed, see response to comment 2. above.

b) *“Attributed to policy changes” – could you please specify which policies?*

The Chinese Coal to Gas policy has been added as an example, the sentence now reads:

“Part of the reduction in China can be attributed to policy and technology changes (e.g. Coal to Gas).”

c) *“Appears to be underestimated” – is this based on comparisons with other OH fields?*

Yes, we found large differences between the TransCom climatology used in this study and the OH fields produced by the TM5-MP model. In the manuscript, we describe an imbalanced prior budget, which is either caused by too low loss or too high production. Potentially more appropriate OH fields are the subject of further investigation in a follow-up study.

d) *The last two sentences are unclear, especially “model’s limited capabilities to capture”; could you please provide more detail?*

The Abstract has been changed accordingly, with the last two sentences, due to length constraints, simplified to:

“In the experiments that include the surface flask measurements, we find strong localized emission increments over Europe and the Sahara, which are traced back to limitations of the model in reproducing point measurements on mountain tops.”

4) *Line 32. Can you please rephrase? What do you mean by ‘dividing them up by source categories.*

The sentence has been rephrased and a clarification was added. It now reads:

“Estimating regional CO emissions and partitioning them by source category (i.e. distinguishing CO from secondary production, fossil fuel combustion, and biomass burning) on a global scale is challenging.”

5) *Line 34. ‘they carry insufficient information’. Please elaborate*

The following elaboration has been added to explain that statement:

“Global remote sensing instruments usually feature very limited vertical resolution and cannot inherently distinguish when, where, and by what process (secondary production, biomass burning, etc.) each observed CO molecule was produced. In addition, the temporal resolution of global remote sensing instruments at a given location is limited to their revisit period (typically on the order of days), which may be insufficient to adequately resolve rapid events, such as biomass burning. The temporal coverage might be further reduced when clouds or other data quality issues make observations temporarily impossible.”

6) *Line 35. What do you mean by ‘incorporating some additional information?’*

Since this was detailed in the following paragraph, the redundant mention has been removed and the sentence now reads:

“However, indirect estimation of CO emission sources from remote sensing data is possible using either bottom-up or top-down approaches.”

7) Line 37-38. 'process that caused the emission is measured'; 'emissions can be extrapolated'. Please rephrase to make it more accurate.

The paragraph has been revised and now reads:

"In bottom-up estimates, the process that produced the emission is modeled based on observations that constrain that process. For example, if the cause of the CO emissions is a wildfire, the emissions can be estimated based on knowledge about the burnt vegetation and the intensity of the fire. Conversely, in top-down estimates, the concentrations that resulted from the emissions are measured and traced back to their source. Again using the example of wildfire CO emissions, their effect in the atmosphere is an elevated CO concentration, that can be observed and then traced back and attributed to its source using atmospheric modeling."

8) Line 42-45. 'direct observations of the source event', 'loose observational requirements'. Please rephrase to make it more accurate. also, 'potentially more elaborate' - why potentially?

The paragraph has been revised. Regarding 'potentially': The term was used because some bottom-up models are very elaborate and some top-down approaches are rather simplistic. Therefore, it is not generally true that top-down approaches always require more elaborate assumptions than bottom-up approaches. The revised paragraph now reads:

"Both approaches are subject to various sources of error. Bottom-up estimates typically require direct observations of the source event (e.g., to have remote sensing information on fire intensity in the case of biomass burning) in addition to certain assumptions about the source itself, such as fuel characterization (ecosystem type, fuel loading, and fuel consumption rates) and emission factors in the case of biomass burning. Top-down estimates do not necessarily require observations of the source event itself, but rather observation(s) of the resulting concentrations at some point in the future are sufficient. However, while the observational requirements of top-down estimates are less strict, they often require a set of more elaborate assumptions for the atmospheric modeling, for example about chemistry and atmospheric transport."

9) Line 48. 'top-down approach in the form of inverse modeling'. are there other forms?

Some researchers distinguish data assimilation and inverse modeling as different categories of top-down approaches. To avoid confusion we have specified that this work is based on a 4DVAR approach and the sentence now reads:

"In this study, we use a top-down approach in the form of four-dimensional variational (4DVAR) inverse modeling, specifically, the state-of-the-art inverse modeling framework TM5-4DVAR."

10) Line 58. 'including information from additional observations'. what do you mean by 'additional'?

We meant 'additional' in the sense of 'not part of the prior already'. The text was updated to clarify: "By incorporating information from additional observations beyond those used to create the a priori emissions, inverse modeling is able to reduce the uncertainties in the a priori emissions that are typically taken from bottom-up inventories."

11) Line 83-96. The zooming capability is a very important point (strength) for this paper which should be highlighted more and taken advantage in extracting full information content of high-resolution datasets to address a science objective. Why would you reduce the observation resolution then? especially that this paper is considering: a) efficacy of high resolution obs, and b) near-real time application.

In the current state of the model, zooming is limited to a resolution of  $1^\circ \times 1^\circ$ , which is already much coarser than what TROPOMI can provide. As outlined above, the references to high-resolution observations + near real-time have been removed, as those were, indeed, not properly investigated in the presented study. The high-resolution observations provided by the TROPOMI instrument are still meaningful in this comparably low-resolution modeling study since the resulting super-observations will have a reduced uncertainty compared to each observation that went into them or to what a lower-resolution instrument could provide.

12) Line 97. *'as a proof of concept'. has there been no inversions using tropomi yet?*

At the time of the initial manuscript submission, there were indeed no inversions using TROPOMI published yet. However, that has changed since, and throughout the manuscript, references to recent studies have been added, e.g. as outlined in the response to comment 3. above

13) Line 117. *Can you please elaborate why CMIP6 emissions are used?*

CMIP(6) emissions are widely used in the modeling community. They are adequate for inversions because they correctly predict where countries are and on what order of magnitude their emissions are. The inversion will correct for any potential mismatches between emissions and TROPOMI CO columns.

14) Line 129-135. *For inversions using full TROPOMI resolution, was the model resolution also change appropriately?*

No, the same zooming setup has been used in all experiments, to ensure comparability of the results. To avoid future confusion, the following sentence was added in L146, where the zooming setup is introduced:

“This zooming setup is used for all inversion experiments presented in this study.”

15) Line 137. *Please elaborate on the rationale for the use of monthly OH from TransCom-CH<sub>4</sub>. Several studies have pointed out issues with using prescribed OH climatology especially for 'regional' inversions.*

OH in the global atmosphere is relatively well buffered, and the Spivakovsky climatology still complies with observed methyl chloroform loss rates. While we are aware of the limitations of the climatology, we are not aware of an alternative that is an objective improvement over the TransCom-OH. As pointed out in recent literature (e.g. Zhao et al. (2019) (<https://doi.org/10.5194/acp-19-13701-2019>) and Naus et al. (2019) (<https://doi.org/10.5194/acp-19-407-2019>)) there is a need for improved OH fields in the community, however, our current understanding and the currently available observations are insufficient to provide that at the moment. More appropriate OH fields are being explored in ongoing investigations. To address the raised concern, a small paragraph has been added to the manuscript (L153ff), which reads:

“Jiang et al. (2017) show that OH is well buffered in the atmosphere on a global scale over the past decades, as indicated by a low month-to-month variability in the methyl chloroform loss rate, and thus the TransCom OH climatology is still considered applicable to recent years, as in e.g. Naus et al. (2022).”

16) Line 173. *please elaborate on 'assimilating multiple datasets with different spatial and temporal resolutions at once and co-sampling of observations across datasets is neither necessary nor detrimental'. what do you mean by 'at once', 'co-sampling' and detrimental to?*

This statement was used to indicate that it is not necessary to only consider those observations across multiple datasets where they happen to occur at similar points in time and space and that it is not detrimental to the inversion results if they do. Since this constitutes unnecessary detail, it has been removed from the manuscript and the text now reads:

“Overall, in 4DVAR, the model is sampled temporally and spatially for each individual data point, and each point provides its own contribution to the cost function. As such, this approach is well suited to simultaneously assimilate multiple datasets with different spatial and temporal resolutions.”

17) Line 191. *'no daily cycles'. while it is consistent with OH, it is not suitable for inversions using 7km data. there's a mismatch in scale.*

Following Naus et al. (2022), who have shown a diurnal cycle in emissions to not be necessary even at 1°x1°, we neglected the daily cycle considering our inversion experiments are done at a resolution of at most 3°x2°, the observational data has at best a daily temporal resolution and the lifetime of CO is multiple weeks to months.

18) Line 230-270. While the use of error correlation lengths is commendable, please elaborate on the scales use in the inversion? How are these calculated/estimated? This could be very useful to the community.

As outlined in that section, the error correlation lengths were taken from previous studies with similar setups in the same model. We agree that having a rigorous approach to calculate them for a given setup would be very useful, however, that was not the focus of this study.

19) Line 296-318. Please rephrase these paragraphs to make it clearer. It is unclear for example what is the exactly spinup period (and pinup inversion) and how was this conducted. For CO, this spinup period can be quite important, especially that there are differences in the model configuration of the initial conditions (incl. more importantly a different OH).

The entire section "Initial conditions, spin-up, and main inversions" has been rearranged and in part rewritten and now reads:

"The initial tracer distribution is an important part of an inversion. Close to the starting date of the inversion period, the initial tracer distribution must fit the total columns and horizontal distribution of the observational datasets reasonably well. If there are significant over- or under-estimations, the emission increments will be dominated by the model's efforts to correct for the offset in the mixing ratios. These additional emissions will mask the true signal of the observations, i.e. by how much the a priori emissions differ from the true emissions. In addition, the initial vertical CO distribution must be realistic, since the CO depletion and transport vary with altitude. Therefore, assuming a too high initial mixing ratio in a layer with low transport and low loss will affect the model for a long time. To minimize this type of error, the period of interest (the year 2018) is split into two separate periods, each with separate inversions, and only the second period is considered for the scientific analysis.

During the first period, a spin-up inversion is performed to harmonize the global distribution of CO mixing ratios in the model with the observational datasets (see Sect. 3). This spin-up inversion is started with tracer fields taken from the TM5-MP chemistry transport model, which employed the MOGUNTIA chemistry scheme. See Myriokefalitakis et al. (2020) and references therein for a detailed description of the model, setup, and chemistry scheme, alongside extensive validation against observational data. In addition to the simulation analyzed and described in Myriokefalitakis et al. (2020), the TM5-MP model has been run with the same settings for a longer period, including 2018. Here, we use the instantaneous concentrations from this longer simulation as initial conditions for the spin-up inversion and monthly chemical budget terms for the secondary source of CO from VOC oxidation. The validations in Myriokefalitakis et al. (2020) have shown that the TM5-MP model generally produces reasonably realistic tracer fields both in terms of vertical and horizontal distributions. However, some offsets to the observations still remain. For CO specifically, Myriokefalitakis et al. (2020) found mixing ratios that were too low in the northern hemisphere and too high in the southern hemisphere. The spin-up inversion in this study is necessary to confidently remove these offsets. In addition, the spin-up inversion facilitates a smooth transition between the different emission datasets used by Myriokefalitakis et al. (2020) in TM5-MP and those used in this study in TM5-4DVAR. While we both use CMIP6 for anthropogenic CO and the same meteorology, they also use CMIP6 for biomass burning, while we use FINN2.5 or GFED4.1s. We use different priors for biomass burning because both inventories (FINN2.5 and GFED4.1s) provide historical data rather than projections for 2018, and inversions benefit greatly from realistic lateral a priori distributions that cannot be obtained from projection data as in CMIP6. Another important difference is the treatment of OH. While their OH is calculated online, we use prescribed OH as described in Sect. 2.1. Overall, harmonizing the mixing ratios modeled in TM5-4DVAR and the observations, especially in remote regions where transport is slow, requires that the model is run over a longer period of time. Therefore, the spin-up inversion is run over several months, from 1 January 2018 to 1 July 2018.

The second period is the main inversion period, which uses the harmonized mixing ratios from the spin-up inversion as initial conditions. The main inversion period spans seven months from 1 June 2018 to 1 January 2019 and leads to the scientifically interesting results presented in Sect. 4. Note

that June is part of both the spin-up and the main inversion periods. This overlap is necessary because emissions near to the end of each inversion period are verified by very few observations. Therefore, the final month of the spin-up inversion is considered as its spin-down period, during which confidence in the generated emissions and the resulting mixing ratios is reduced. Similarly, the final month of the main inversions, December 2018, should be considered as their spin-down period. The duration of this spin-down period was chosen based on the lifetime of CO of about two months (Raub et al., 1991; Holloway et al., 2000). Hence, a snapshot of the mixing ratios from the final iteration of the spin-up inversion of 1 June 2018 is used as initial conditions for the main inversion. By using these mixing ratios from the spin-up inversion, which are already harmonized to the observations as initial conditions, no further spin-up is required for the main inversions and their June results can already be trusted.”

20) *Figure 2. Nice figure to better understand the elaborate gridding strategy. Still not sure though that this is appropriate for near real-time applications.*

The term near real-time has been removed from the manuscript, see response to comment 2. above.

21) *Line 517-524. Nice discussion on the limitation. It still begs the question (same as above comment) if this elaborate weighting strategy is appropriate.*

We agree that the described method will not be useful for near real-time applications as is, however, further developments on this basis are under investigation, some of which are aiming at significantly reducing the computational cost.

22) *Table 3. Very helpful to have.*

Thank you for the comment.

23) *The manuscript's honest discussion of inversion increments and identified issues is commendable, yet it risks obscuring key findings and blurring the clarity of its objectives. Reorganizing the structure and sharpening the focus would enhance clarity and impact.*

The manuscript has been reorganized as suggested to improve its structure and sharpen the focus.